Dissecting Real World Systems with the RIXS Scalpel

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We have built a reaction cell for catalytic ethylene epoxidation over silver operating at atmospheric pressures of ethylene/oxygen gas mixtures. I will present results from our in situ x-ray spectroscopy studies of this system under various sample, gas mixture, and temperature conditions. The reaction cell can contain either a compressed pellet of Ag powder or an evaporated Ag film on a silicon nitride membrane. In both cases, the silicon nitride membrane allows x-rays to penetrate into the silver where special oxygen sites (e.g. electro- and nucleophilic) form under elevated temperatures (up to 250° C) and are the key to the catalytic partial oxidation of ethylene. X-ray absorption spectroscopy (XAS) and resonant inelastic x-ray spectroscopy (RIXS) reveal how the oxygen sites evolve under different reaction conditions. Although XAS spectra are superpositions of gas phase and incorporated oxygen, there is a clear signature from the latter. Moreover, the energy selectivity of RIXS is employed as a powerful tool to project the specific sites that are of interest. Our study fills the long standing pressure gap that has plagued realistic studies of this system and points the way for future in operando studies on similar systems.